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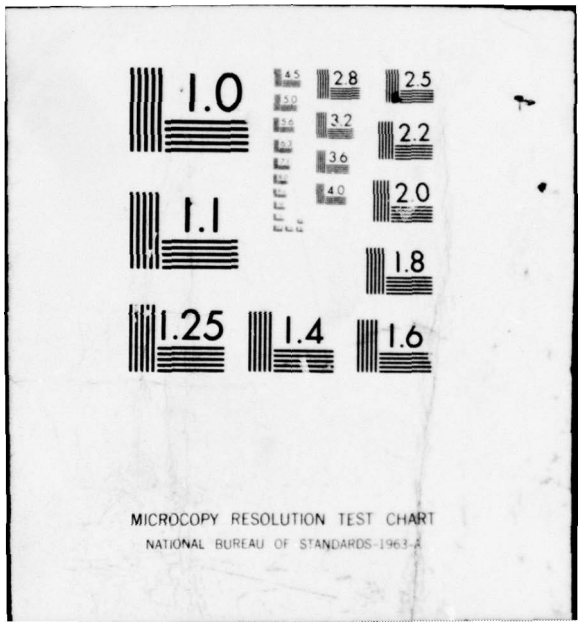
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Research and Development Technical Report

ECOM-4437

DESIGN OF A NOZZLE BEAM TYPE METAL VAPOR SOURCE

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October 1976

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US ARMY ELECTRONICS COMMAND FORT MONMOUTH, NEW JERSEY 07703

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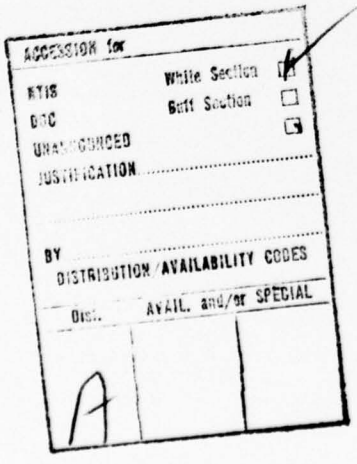

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SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

REPORT DOCUMENTATION PAGE

READ INSTRUCTIONS BEFORE COMPLETING FORM

(14) 1. REPORT NUMBER: ECOM-4437

2. GOVT ACCESSION NO.

(9) Research and development technical rept.

3. RECIPIENT'S CATALOG NUMBER

(6) 4. TITLE (and Subtitle): Design of a Nozzle Beam Type Metal Vapor Source.

5. TYPE OF REPORT & PERIOD COVERED

6. PERFORMING ORG. REPORT NUMBER

(10) 7. AUTHOR(s): Ronald P. Andres*

8. CONTRACT OR GRANT NUMBER(s)

9. PERFORMING ORGANIZATION NAME AND ADDRESS
US Army Electronics Command
ATTN: DRSEL TL-MF
Fort Monmouth, N.J. 07703

10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS
1S7 62705 AH94 F3 01

11. CONTROLLING OFFICE NAME AND ADDRESS
US Army Electronics Command
ATTN: DRSEL TL-MF
Fort Monmouth, N.J. 07703

(11) 12. REPORT DATE: October 1976

13. NUMBER OF PAGES: 17

14. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office)

(12) 22p.

15. SECURITY CLASS. (of this report): Unclassified

15a. DECLASSIFICATION/DOWNGRADING SCHEDULE

16. DISTRIBUTION STATEMENT (of this Report)

Statement A.
Approved for public release; distribution unlimited.

17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)

18. SUPPLEMENTARY NOTES

* Prof. Andres, Princeton U. performed the basic work upon which this paper is based under prior Scientific Svcs Program Contr DAHC04-72-A-0001 - Task 75-7.

This report appeared in the Proceedings of the 30th Annual Symposium on Frequency Control, 1976.

19. KEY WORDS (Continue on reverse side if necessary and identify by block number)

quartz-resonators	evaporation-sources	effusive-flow
frequency-control	nozzle-beams	jet-flow
vacuum-deposition	molecular-beams	
gold	collimated-beams	

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

A prototype design of a directional high flux source for use in vapor deposition of electrode materials during fabrication of precision quartz crystal resonators is presented. The design is based on the theory and technology of nozzle beams. This nozzle beam type source is conceived: (1) to permit large deposition rates with minimum wastage of electrode materials, (2) to operate in high vacuum, (3) to emit vapor in a horizontal direction, thereby permitting the use of a pair of sources to plate both sides of a substrate simultaneously,

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and (4) to operate for extended periods without requiring frequent breaking of the vacuum in order to replenish the source. It is estimated that this design can operate at a deposition rate equal or above that of a conventional evaporation type source with less than one percent of the wastage of electrode material experienced with a conventional source.

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Introduction

A critical step in the fabrication of ultra high precision quartz crystal resonators is the vacuum deposition of the electrode material. In order to minimize stresses that could cause aging, both sides of the resonator are plated simultaneously and at equal rates so that the final thicknesses of the electrodes are approximately equal. In order to minimize aging due to mass transfer, the electrode material is of high purity and is deposited rapidly under high vacuum conditions so as to minimize sorption of contaminants.

These requirements place rather stringent performance requirements on the metal vapor source used in the vacuum plating operation. Ideally it should minimize wastage of expensive electrode material (such as gold), should be capable of operation over a wide range of controlled deposition rates, should be compatible with high vacuum operation, should not require frequent maintenance, and should permit simultaneous plating of both sides of a substrate.

These requirements led Drs. E. Hafner and J. Vig of the U.S. Army Electronics Command, Fort Monmouth, New Jersey, to investigate the possibility of using molecular beam techniques to develop a new highly directional vapor deposition source [1]. Their concept was a source such as is shown schematically in Figure 1. This source is divided into two chambers interconnected by means of an aperture. The first of these chambers (source chamber) is held at a high temperature so as to maintain the vapor pressure of the evaporant at a high value. The second chamber (collimation chamber) is maintained at the melting point of the evaporant thereby maintaining a relatively low vapor pressure. Because of the pressure differential between the two chambers, a vapor flow is established through the source aperture. A portion of this flow passes through a collimation aperture and plates the substrate. By far the largest fraction of the flow, however, strikes the walls of the collimation chamber,

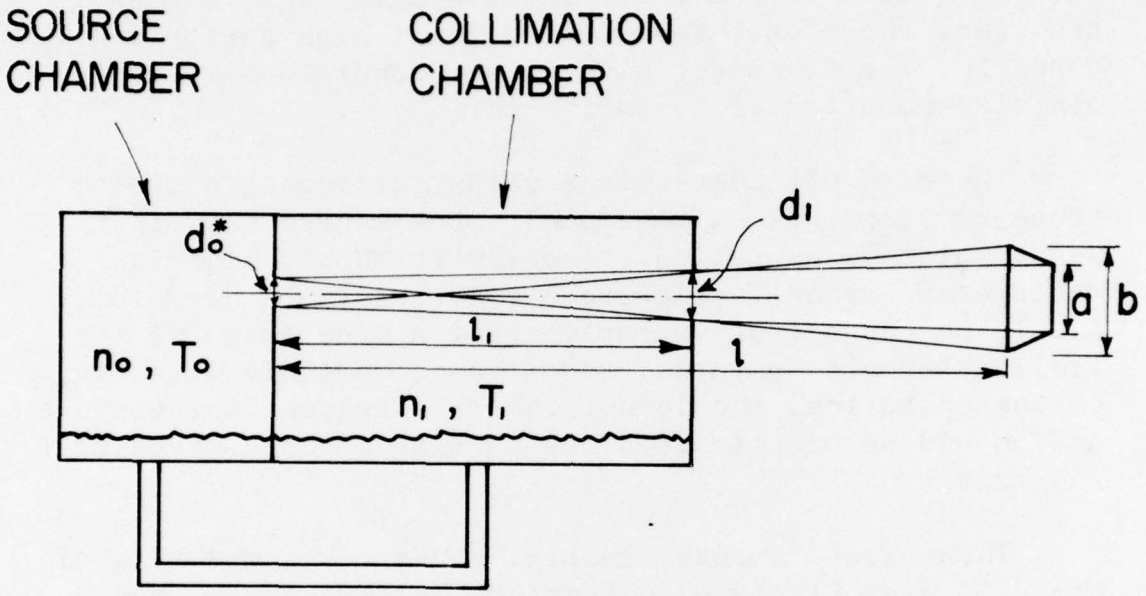

Figure 1. Schematic of Nozzle Beam Type Source

condenses, and is recycled to the source chamber by means of a liquid transfer line. The great advantage of such a design over conventional evaporation sources is that the major part of the evaporant flow not needed to plate the substrate is recycled and not wasted. The need for high deposition rates leads to the requirement that the flow from the source chamber be at a relatively high density. Such unconstrained expansion of a vapor from a high pressure source through an orifice into a vacuum is termed a nozzle or free jet flow.

This paper sets forth general design considerations for such a nozzle beam type source and a theoretical model for its operation. Experiments run with several source configurations are reported. Finally a prototype design is presented and discussed.

General Design Considerations

The flow distribution from a circular orifice into a region of vacuum can be theoretically predicted both in the limit of vanishing source density (i.e., large mean free path in the source relative to the diameter of the source aperture) and in the limit of high source density (i.e., small mean free path relative to aperture diameter).

In the first case we have the classical expressions for effusive flow:

$$f_o = (0.282)n_o\left(\frac{2kT_o}{m}\right)^{1/2}\left(\frac{\pi d_o^2}{4}\right) \tag{1}$$

and

$$I(\theta=0,\ell) = f_o\left(\frac{1}{\pi\ell^2}\right) , \tag{2}$$

where

f_o = total source flow (molecules/sec)

n_o = source density (molecules/cc)

T_o = source temperature (°K)

k = Boltzmann's constant (erg/°K)

m = molecular weight of vapor molecule (gm/molecule)

d_o = source aperture diameter (cm)

$I(\theta=0,\ell)$ = centerline flux intensity (molecules/cm sec)

ℓ = distance from source aperture (cm).

In this effusive limit the flux intensity, $I(\theta,\ell)$, varies as the cosine squared of the angle, θ, measured with respect to the centerline of the flow. Thus, in the region about $\theta = 0$ the flux of molecules is: (1) practically constant with θ, (2) proportional to the total source flow, f_o, and (3) inversely proportional to the distance, ℓ, squared.

Sherman and Ashkenas [2] have analyzed the structure of the supersonic free jet flow that exists in the limit of high source density. Making use of their results yields simple expressions for total source flow and centerline intensity which have the form:

$$f_o = (0.513)n_o\left\{\frac{2kT_o}{m}\right\}^{1/2}\left\{\frac{\pi d_o^{\,2}}{4}\right\} \tag{3}$$

$$I(\theta=0,\ell) = (1.85)f_o\left\{\frac{1}{\pi\ell^2}\right\} \tag{4}$$

The numerical constants in Eqns. (3) and (4) are for the case of a monatomic vapor. The flux intensity, $I(\theta,\ell)$, varies in this high density limit as

$$I(\theta,\ell) \simeq I(\theta=0,\ell)\cos^2\left\{\frac{\pi\theta}{2\phi}\right\} \qquad (5)$$

where $\phi = 1.36$ radians for the case of a monatomic vapor. Thus, in the region about $\theta = 0$ the flux of molecules is again: (1) practically constant with θ, (2) proportional to the total source flow, f_o, and (3) inversely proportional to the distance, ℓ, squared. In the high density or nozzle flow limit, however, the centerline flux intensity for a given source flow is nearly twice what it would be if effusive conditions obtained. Furthermore, the source flow and thereby the centerline intensity of a nozzle type source can be orders of magnitude greater than the effusive values for a given size aperture thereby permitting much higher vapor deposition rates.

In most plating operations the distance, ℓ, is taken to be fairly large so as to minimize the effects of thermal radiation from the source. The actual solid angle about $\theta = 0$ that is subtended by the substrate is a small fraction of the total solid angle of the flow. Thus, only a small fraction of the total vapor passing through the source aperture is deposited on the substrate. The remainder of the source flow deposits onto various collimators or masks used to define the actual area on the substrate that is to be plated and onto the walls of the vacuum system. While this material can in principle be recovered for eventual reuse, such recovery involves breaking open the vacuum system and substantial repurification. Ideally one would like to continuously recycle the material that is not deposited on the substrate. It is this concept that forms the basis of a nozzle beam type source.

The elements of such a source are schematically pictured in Figure 1. A directed beam of vapor molecules is collimated from the total flow passing through the source aperture and this collimated beam is used to plate the substrate. The remaining portion of the vapor condenses on the walls of the chamber containing the collimation aperture. This collimation chamber is then either maintained at a temperature above the melting point of the material being used so that the material continuously flows back into the source chamber or it is periodically heated to melt and recycle the condensate.

As long as n_1 is kept low enough so that the vapor molecules flowing through the source aperture are not appreciably scattered by the background density in the collimation chamber, the source flow will become free molecular at some point and the intensity distribution in the collimated beam will be roughly that from a virtual free molecular source situated at the position of the source aperture [3]. In the case of effusive flow the diameter of this virtual source, d_o^*, equals the actual aperture diameter, d_o. In the nozzle or high density limit the diameter of the virtual source is a multiple, α, of the actual aperture diameter, where α depends on the product $n_o d_o$ (increasing slowly as $n_o d_o$ increases). The parameter, α, is always greater than unity but is typically less than four for flows in which nucleation and condensation of the vapor do not take place [3].

The intensity distribution from such a collimated free molecular source is shown schematically in Figure 1. The intensity profile consists of two regions: (1) a central portion characterized by a diameter, a, in which the intensity is that of the uncollimated flow and (2) an annular region characterized by an outer diameter, b, in which the intensity falls to essentially zero. Straightforward geometrical considerations yield the following expressions for a and b:

$$a = d_1 \frac{\ell}{\ell_1} + d_o^* \left(1 - \frac{\ell}{\ell_1} \right) \tag{6}$$

$$b = d_1 \frac{\ell}{\ell_1} + d_o^* \left(\frac{\ell}{\ell_1} - 1 \right) \tag{7}$$

The only part of such a collimated beam that is useful for uniform plating of a substrate is the central portion. The diameter of this constant flux region can be designed, however, to be equal to or slightly greater than the diameter of the substrate area that is to be plated. The essential feature of such a collimated beam source is that only that fraction of the vapor flowing through the collimation aperture that is directed into the annular region with inner diameter "a" and outer diameter "b" need be wasted in contrast to the usual case of an uncollimated source when everything passing through the source aperture that is not directed onto the substrate is wasted. Even the small fraction of the flow that is wasted can be minimized by keeping d_o^*/d_1 small and/or by keeping ℓ/ℓ_1 close to unity.

Gold Vapor Source

It is possible, using relevant physical data [4] and Eq. (3), to evaluate f_o for a nozzle type gold vapor source. The results of such a calculation are plotted in Figure 2 as a function of source temperature, T_o. A standard aperture diameter, d_o, of 1 mm was arbitrarily assumed in these calculations. The flow through any other aperture can be found by multiplying f_o from Figure 2 by the square of the actual diameter in mm. Two sets of units in which to express f_o are used. The left hand scale reads in molecules/second while the right hand scale reads in milliliters liquid gold/second.

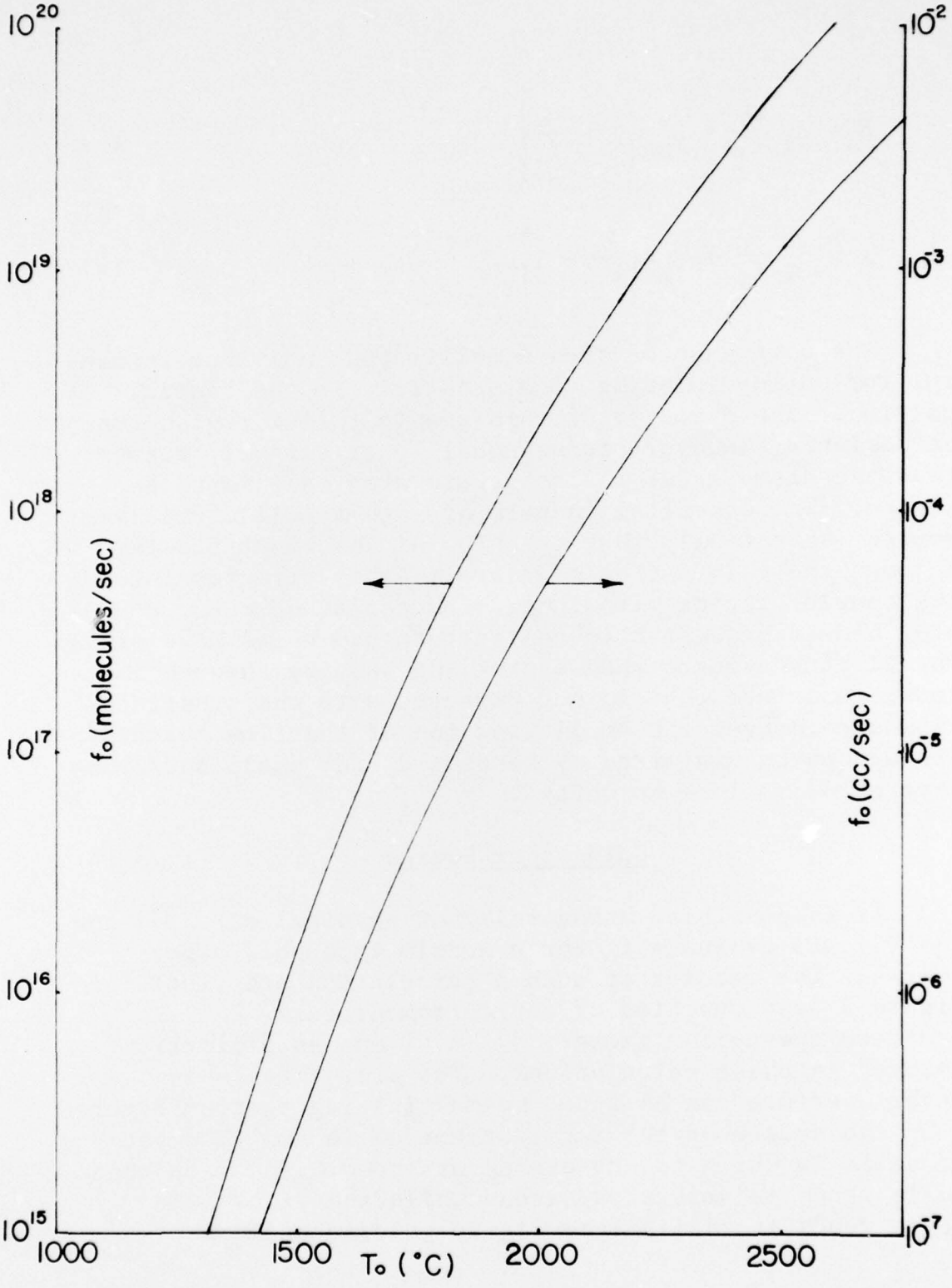

Figure 2. Gold Flow Through One Millimeter Diameter Aperture

The ideal centerline intensity of a nozzle beam type gold vapor source can now be obtained through use of Eq. (4) and values of f_o obtained from Figure 2. As an example of the order of magnitude of this flux, assume a standard arrangement in which d_o = 1 mm and ℓ = 10 cm. Expressing the flux of gold vapor as an equivalent deposition rate, the deposition rate for this configuration in Å/sec equals 10^{-17} times f_o expressed in mol/sec.

A rate of 0.01 Å/sec is achieved when $T_o \approx 1300°C$. A deposition rate of 10 Å/sec requires a source chamber temperature of 1900°C. High deposition rates, therefore, restrict the material of construction for a gold vapor source to one of the refractory metals.

The temperature of the collimation chamber for the case of a gold vapor source is set by the melting point of gold, 1063°C. At this temperature there are a number of oxide ceramics that can be used as materials of construction. In particular, high purity Al_2O_3 seems attractive. It exhibits negligible chemical interaction with Au, is an electrical insulator, is easily fabricated into practically any shape, is tolerant of thermal shock, and is inexpensive[2].

Experimental Considerations

The primary difficulties in achieving a practical source based on the foregoing ideas arise from: (1) the high temperatures required and (2) the need to move the liquid evaporant against a pressure differential from the collimation chamber to the source chamber. A number of experiments were carried out to determine a suitable solution to these problems. In all of these experiments Cu was used as the evaporant. The material of choice for the collimation chamber (crucible) was high purity Al_2O_3 and for the source chamber was tungsten; although in some of the experiments lava was substituted for Al_2O_3 and tantalum was substituted for tungsten because their availability and lower cost permitted a larger range of experiments to be performed.

The easiest way to heat a refractory metal object to a high temperature is by electrical resistance heating. In this case the best geometry for the object is that of a cylinder with a small O.D. Thus, in all of the experiments the configuration of the source chamber was that of a long thin cylindrical tube, closed at one end and communicating with the fluid reservoir via the opposite open end. The source aperture was drilled into the side of the tube.

The only feasible way to transfer the liquid evaporant appears to be by means of capillary forces or by means of a differential liquid head. The contact angle of liquid gold on a clean tungsten surface in vacuum is less than 90° and decreases with increasing temperature [5]. Thus, it is expected that for this system surface tension forces will tend to draw liquid evaporant into the source tube. In all of the experiments with Cu, however, the opposite phenomenon was observed, i.e. there was a decided capillary lowering of the liquid level in the tube. A contact angle greater than 90° was also observed at the Cu-Al_2O_3 interface.

The first experimental configuration studied is shown schematically in Figure 3. The collimation chamber had a helical groove machined on the outside. Tungsten wire (0.015" diam.) was wound in this groove and was used to electrically heat the crucible. This technique proved very successful and was improved in later models by transforming the groove into one with a flat cross section in which 1/16" wide by 0.005" thick tantalum ribbon could be wound and by increasing the thickness of the crucible's bottom to assure a more uniform inside temperature. The source chamber was a 1/8" O.D. tantalum tube with a wall thickness of 0.010". Electrical current to heat the tube was supplied by means of a stainless steel clamp at the closed upper end and by means of a 1/8" O.D. tungsten rod immersed into the pool of liquid metal at the bottom of the crucible.

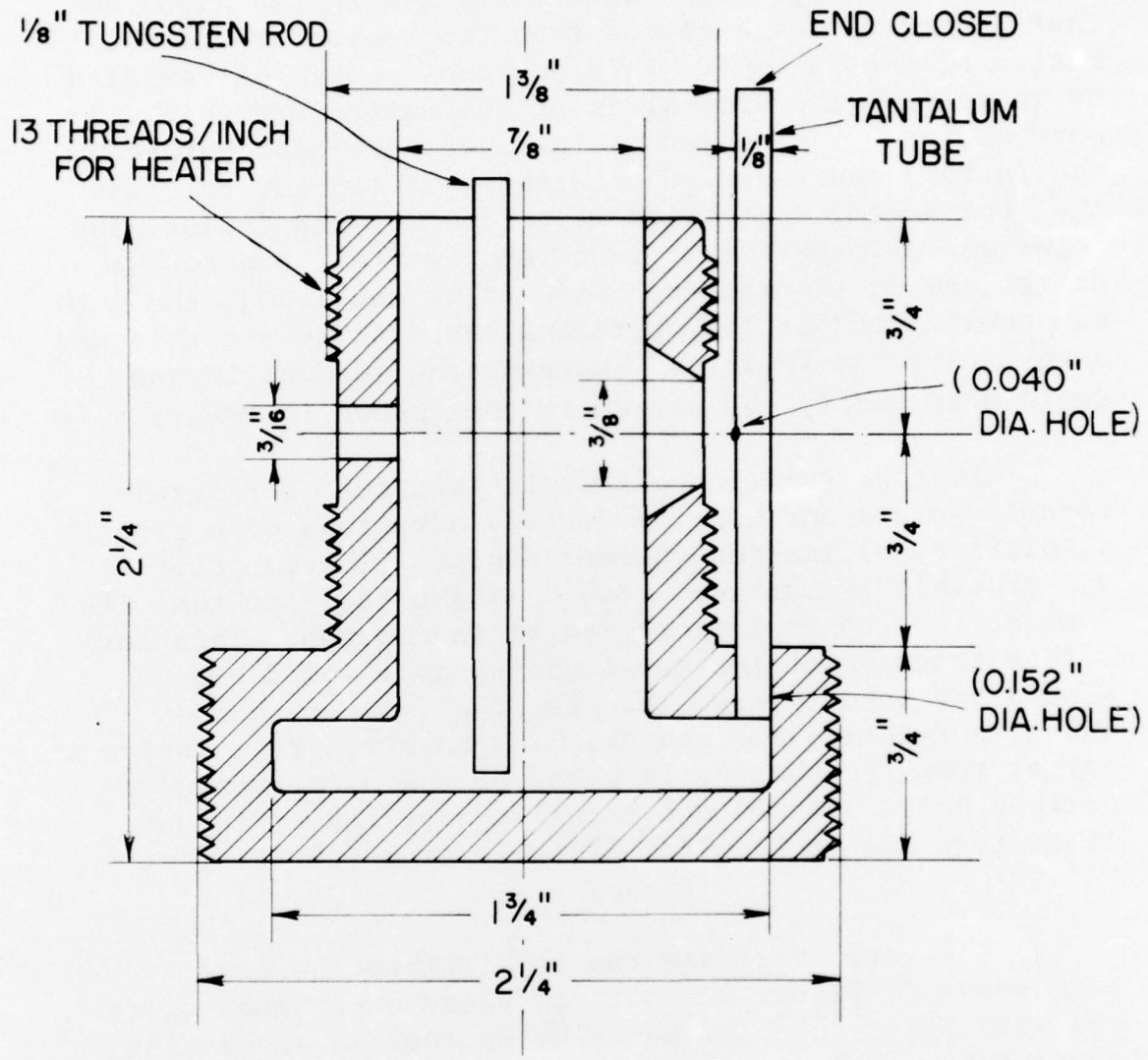

Figure 3. Experimental Nozzle Beam Type Source

Several attempts were made to test the performance of this design. None of these runs were successful in producing a copper beam. They served to point out a number of serious flaws in the design. First, copper exhibited a relatively large contact angle with the Al_2O_3 so that liquid Cu was excluded from the region around the bottom of the Ta tube. This could of course be remedied by increasing the dimensions of the entire crucible. A more serious flaw, however, involved the joint between the Ta tube and the Al_2O_3 crucible. After several runs the tube became embrittled at this joint and failed. The joint was a force fit at room temperature. Due to the difference in thermal expansion of Ta and Al_2O_3, the tube was constricted at its operating temperature and this may have caused the failure. Whatever the reason, it was decided to modify the source to that shown in Figure 4.

This configuration eliminates the need for metal-ceramic joints and has the decided advantage of a greatly simplified collimation chamber design. The interior of the crucible is machined with a slight taper so that its I.D. at the top is larger than at the bottom. This permits extraction of the metal charge as a solid block should the need arise. The prototype source chamber shown in Figure 4, i.e., a small O.D. tungsten tube inside a larger tungsten sheath, is based on the experiments described below but has not actually been tested in our laboratory due to the lack of facilities for its fabrication.

Two experiments were run to simulate the source design shown in Figure 4. Both of these experiments were run with inner tubes of tantalum encased in an alumina outer tube. The first experiment involved a 1/8" O.D. tantalum tube with a wall thickness of 0.010" encased in a 10 mm O.D., 6 mm I.D., alumina tube. The alumina tube was originally closed at one end, and a hole was drilled through this end to permit passage of the tantalum tube. The joint was a force fit at room temperature.

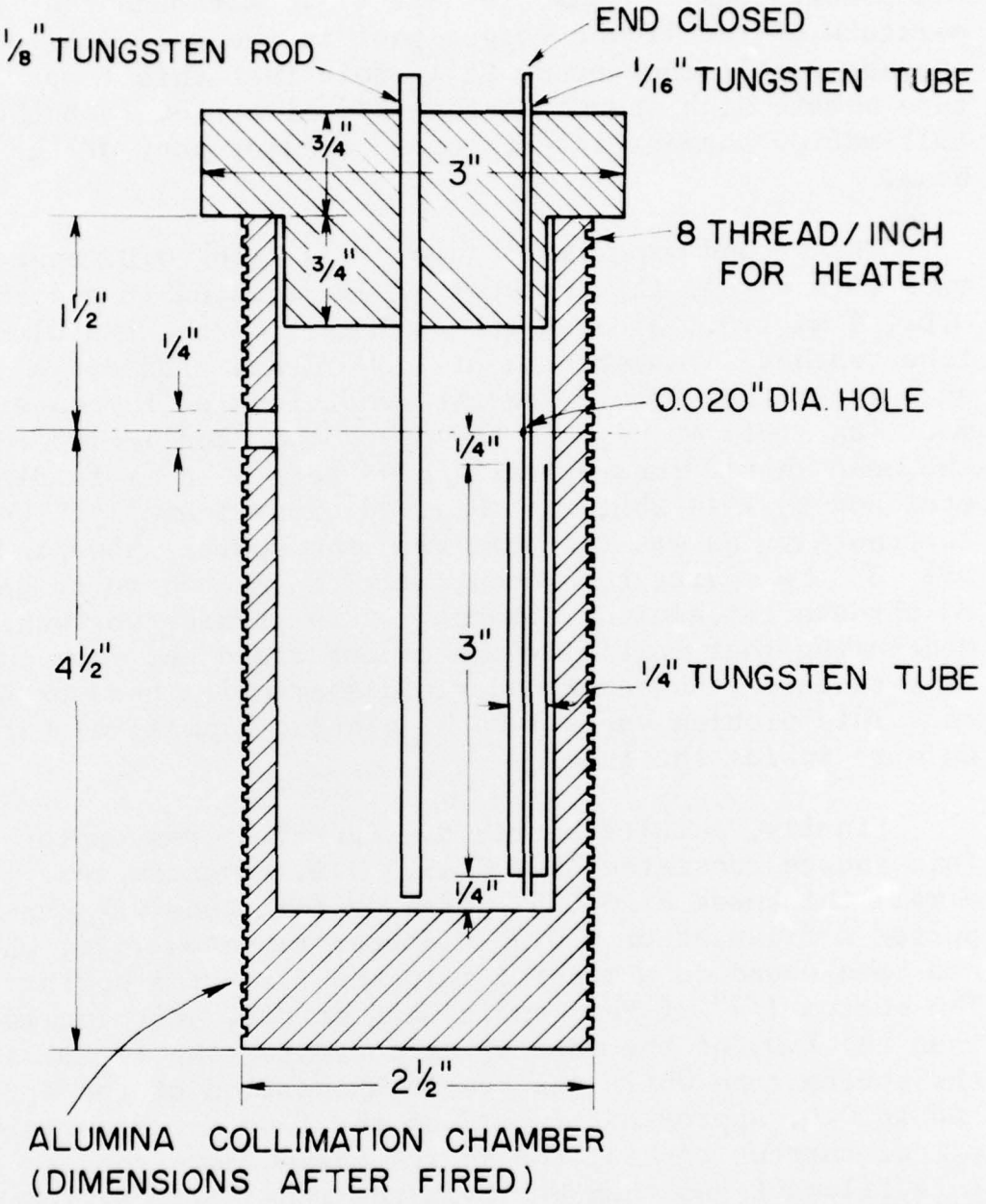

Figure 4. Prototype Nozzle Beam Type Source

This source produced an intense copper beam. The temperature of the tantalum tube, however, did not rise much above that of the crucible and the spatial distribution of the beam, while directed, did not agree with the predictions of Eqns. (6) and (7). Although the temperature of the liquid copper pool in the collimation chamber wasn't monitored, it is felt that this temperature became high enough so that effusive flow from the collimation chamber itself formed a major part of the beam.

The second experiment involved a 1/16" O.D. tantalum tube with a wall thickness of 0.005" encased in a 4 mm O.D., 2 mm I.D., alumina tube. This time the tantalum tube reached a temperature of 2,000°C and produced a sharply defined beam. For the conditions of the experiment (d_o = 0.5 mm, ℓ_1 = 5 cm, d_1 = 6 mm, and ℓ = 15 cm) the beam should, theoretically, have been $\simeq$ 18 mm in diameter and this is what was observed experimentally. Unfortunately, as was the case with the source shown in Figure 3, the source tube eventually cracked where it passed through the alumina sheath. It is perhaps worthwhile mentioning that initially the copper would not flow into this tantalum tube even under a differential head of 4 cm. This problem was solved by placing a small amount of Cu wire inside the tube.

Finally, a third source configuration was explored. This source consisted of a 0.195" O.D. tungsten tube with a wall thickness of 0.005". Inside this tube was supported a filament of 0.015" diameter tungsten wire, which had been wound on a mandril into the form of a spring. The bottom 1/4" of this spring had an O.D. just smaller than the I.D. of the tube so as to center the filament in the source tube while the remaining portion of the spring had an O.D. approximately 1/3 of the tube's I.D. Electrical current to heat the source passed down through this filament and then back via the tube. It was possible to control the source chamber temperature at above 2000°C with this arrangement. Unfortunately, the portion of the tube submerged in the copper pool remained at a relatively low temperature and no measureable copper beam was produced.

As was the case with the other experiments, the dimensions of the components used in this source were dictated primarily by their availability in our laboratory. The liquid level inside the source tube for this configuration will, in general, lie below that of the liquid surrounding the tube. Thus, the tube wall in the region of the liquid-vapor interface will be near that of the surrounding liquid (i.e. approximately the melting temperature of evaporant). The temperature of the liquid near the filament will, on the other hand, be much higher, approaching that of the filament itself. Accurate modeling of this situation is quite difficult but it seems that there should be situations in which an appreciable net flux of evaporant into the tube exists. Because of its relatively simple construction, this configuration should be studied further.

Proposed Design

A suggested prototype nozzle beam type source for use in vapor deposition is shown schematically in Figure 4. This design makes use of the theoretical and experimental considerations described in the previous sections. The dimensions of the collimation chamber in Figure 4 were arrived at by assuming the substrate is located at a distance of 20 cm from the source orifice with a diameter of about 2.5 cm. Under different situations the size of the collimation aperture can be altered following Eqns. (6) and (7) and the earlier discussion.

The Al_2O_3 collimation chamber employed in this design has proved simple to operate and quite reliable. Because of its large size and relatively high operating temperature, it is necessary to provide radiation shielding around this chamber so as to not raise the temperature of the substrate. It may prove in practice that the best way to operate is to actually have the collimation chamber near room temperature during substrate plating and to heat this chamber to recycle evaporant only after the plating operation is over. Whatever turns out to be the case, a thermocouple should be placed inside a closed end alumina tube and immersed in the metal pool in this chamber to monitor its temperature.

The source chamber in Figure 4 has been shown to be feasible in concept but has not been constructed or operated. Probably further experiments with a tantalum source having this configuration should be carried out before constructing one from tungsten. Certainly more work has to be done before the design of this critical part of the source is finalized (see discussion in section on experimental considerations).

This source should exhibit only a very small fraction of the wastage of electrode material experienced with conventional sources. It is estimated that such a nozzle beam type source can operate with deposition rates equal or above that of conventional evaporation sources. At the same time this source is capable of very low, controlled rates of deposition. A source containing two separate source chambers with different size apertures would yield an even larger range of controlled plating rates.

Acknowledgments

I want to thank Drs. E. Hafner and J. Vig for numerous helpful discussions and suggestions throughout this work. Finally, I want to acknowledge the aid of J. Bittner who built the experimental models and of Y. Wong who ran many of the experiments.

Footnotes

1 This work was supported through the U.S. Army Research Office by the U.S. Army Electronics Command, Fort Monmouth, New Jersey.

2 Fabrication with alumina is achieved by starting with green material, which is easily machined, and then firing. Shrinkage of about a factor of 1.25 occurs on firing. The Al_2O_3 collimation chambers used in the present work were fabricated by Western Gold and Platinum Co., 205 Oraton Street, Newark, New Jersey, 07104.

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